5,6-Dialkyl-7-aminotriazolopyrimidines, their preparation and their use for controlling harmful fungi, and compositions comprising these compounds

Description

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The present invention relates to 5,6-dialkyl-7-aminotriazolopyrimidines of the formula I

$$N - N$$
 R^1
 R^2

in which the substituents are as defined below:

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 R^1 is C_1 - C_5 -alkyl or C_1 - C_{10} -alkoxy- C_1 - C_{10} -alkyl,

 R^2 is C_5 - C_{12} -alkyl,

where R¹ and/or R² may be substituted by one to three of the following groups:

cyano, nitro, hydroxyl, C₃-C₆-cycloalkyl, C₁-C₆-alkylthio or NR^aR^b;

 R^a , R^b are hydrogen or C_1 - C_6 -alkyl.

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Moreover, the invention relates to processes for preparing these compounds, to compositions comprising them and to their use for controlling phytopathogenic harmful fungi.

5,6-Dialkyl-7-aminotriazolopyrimidines are proposed in a general manner in GB 1 148 629. Individual fungicidally active 5,6-dialkyl-7-aminotriazolopyrimidines are known from EP-A 141 317. However, in many cases their activity is unsatisfactory. Based on this, it is an object of the present invention to provide compounds having improved activity and/or a wider activity spectrum.

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We have found that this object is achieved by the compounds defined at the outset. Furthermore, we have found processes and intermediates for their preparation, compositions comprising them and methods for controlling harmful fungi using the compounds I.

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The compounds of the formula I differ from those in the abovementioned publications by the specific arrangement of the substituent in the 5-position of the triazolopyrimidine skeleton.

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Compared to the known compounds, the compounds of the formula I are more effective against harmful fungi.

The compounds according to the invention can be obtained by different routes. Advantageously, the compounds according to the invention are obtained by reacting substituted ß-keto esters of the formula II with 3-amino-1,2,4-triazole of the formula III to give 7-hydroxytriazolopyrimidines of the formula IV. The groups R¹ and R² in formulae II and IV are as defined for formula I and the group R in formula II is C₁-C₄-alkyl; for practical reasons, preference is given here to methyl, ethyl or propyl.

The reaction of the substituted ß-keto esters of the formula II with the aminoazoles of the formula III can be carried out in the presence or absence of solvents. It is advantageous to use solvents to which the starting materials are substantially inert and in which they are completely or partially soluble. Suitable solvents are in particular alcohols, such as ethanol, propanols, butanols, glycols or glycol monoethers, diethylene glycols or their monoethers, aromatic hydrocarbons, such as toluene, benzene or mesitylene, amides, such as dimethylformamide, diethylformamide, dibutylformamide, N,N-dimethylacetamide, lower alkanoic acids, such as formic acid, acetic acid, propionic acid, or bases, such as alkali metal and alkaline earth metal hydroxides, alkali metal and alkaline earth metal oxides, alkali metal and alkaline earth metal hydrides, alkali metal amides, alkali metal and alkaline earth metal carbonates and also alkali metal bicarbonates, organometallic compounds, in particular alkali metal alkyls, alkylmagnesium halides and also alkali metal and alkaline earth metal alkoxides and dimethoxymagnesium, moreover organic bases, for example tertiary amines, such as trimethylamine, triethylamine, triisopropylethylamine, tributylamine and Nmethylpiperidine, N-methylmorpholine, pyridine, substituted pyridines, such as collidine, lutidine and 4-dimethylaminopyridine, and also bicyclic amines and mixtures of these solvents with water. Suitable catalysts are bases, such as those mentioned above, or acids, such as sulfonic acids or mineral acids. With particular preference, the reaction is carried out without solvent or in chlorobenzene, xylene, dimethyl sulfoxide or N-methylpyrrolidone. Particularly preferred bases are tertiary amines, such as triisopropylethylamine, tributylamine, N-methylmorpholine or N-methylpiperidine. The temperatures are from 50 to 300°C, preferably from 50 to 180°C, if the reaction is carried out in solution [cf. EP-A 770 615; Adv. Het. Chem. 57 (1993), 81ff].

The bases are generally employed in catalytic amounts; however, they can also be employed in equimolar amounts, in excess or, if appropriate, as solvent.

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In most cases, the resulting condensates of the formula IV precipitate from the reaction solutions in pure form and, after washing with the same solvent or with water and subsequent drying, they are reacted with halogenating agents, in particular chlorinating or brominating agents, to give the compounds of the formula V in which Hal is chlorine or bromine, in particular chlorine. The reaction is preferably carried out using chlorinating agents such as phosphorus oxychloride, thionyl chloride or sulfonyl chloride at from 50°C to 150°C, preferably in excess phosphorus oxytrichloride at reflux temperature. After evaporation of the excess phosphorus oxytrichloride, the residue is treated with ice-water, if appropriate with addition of a water-immiscible solvent. In most cases, the chlorinated product isolated from the dried organic phase, if appropriate after evaporation of the inert solvent, is very pure and is subsequently reacted with ammonia in inert solvents at from 100°C to 200°C to give the 7-amino-triazolo[1,5-a]pyrimidines. This reaction is preferably carried out using a 1- to 10-molar excess of ammonia, under a pressure of from 1 to 100 bar.

The novel 7-aminoazolo[1,5-a]pyrimidines are, if appropriate after evaporation of the solvent, isolated as crystalline compounds, by digestion in water.

The ß-keto esters of the formula II can be prepared as described in Organic Synthesis Coll. Vol. 1, p. 248, and/or they are commercially available.

Alternatively, the novel compounds of the formula I can be obtained by reacting substituted acyl cyanides of the formula VI in which R¹ and R² are as defined above with 3-amino-1,2,4-triazole of the formula III.

The reaction can be carried out in the presence or absence of solvents. It is advantageous to use solvents to which the starting materials are substantially inert and in which they are completely or partially soluble. Suitable solvents are in particular alcohols, such as ethanol, propanols, butanols, glycols or glycol monoethers, diethylene glycols or their monoethers, aromatic hydrocarbons, such as toluene, benzene or mesitylene, amides, such as dimethylformamide, diethylformamide, dibutylformamide, N,N-dimethylacetamide, lower alkanoic acids, such as formic acid, acetic acid, propionic acid, or bases, such as those mentioned above, and mixtures of these solvents with water. The reaction temperatures are from 50 to 300°C, preferably from 50 to 150°C, if the reaction is carried out in solution.

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The novel 7-aminotriazolo[1,5-a]pyrimidines are, if appropriate after evaporation of the solvent or dilution with water, isolated as crystalline compounds.

Some of the substituted alkyl cyanides of the formula VI required for preparing the 7-aminoazolo[1,5-a]pyrimidines are known, or they can be prepared by known methods from alkyl cyanides and carboxylic acid esters using strong bases, for example alkali metal hydrides, alkali metal alkoxides, alkali metal amides or metal alkyls (cf.: J. Amer. Chem. Soc. 73, (1951), p. 3766).

If individual compounds I cannot be obtained by the routes described above, they can be prepared by derivatization of other compounds I.

If the synthesis yields mixtures of isomers, a separation is generally not necessarily required since in some cases the individual isomers can be interconverted during work-up for use or during application (for example under the action of light, acids or bases). Such conversions may also take place after use, for example in the treated plants when plants are treated, or in the harmful fungus to be controlled.

In the definitions of symbols given in the above formulae, collective terms were used which are generally representative of the following substituents:

halogen: fluorine, chlorine, bromine and iodine:

alkyl: saturated straight-chain or mono- or dibranched hydrocarbon radicals having 1 to 4 or 5 to 12 carbon atoms, for example C₁-C₆-alkyl such as methyl, ethyl, propyl, 1-methylethyl, butyl, 1-methylpropyl, 2-methylpropyl, 1,1-dimethylethyl, n-pentyl, 1-methylbutyl, 2-methylbutyl, 3-methylbutyl, 2,2-dimethylpropyl, 1-ethylpropyl, hexyl, 1,1-dimethylpropyl, 1,2-dimethylpropyl, 1-methylpentyl, 2-methylpentyl, 3-methylpentyl, 4-methylpentyl, 1,1-dimethylbutyl, 1,2-dimethylbutyl, 1,3-dimethylbutyl, 2,2-dimethylbutyl, 2,3-dimethylbutyl, 3,3-dimethylbutyl, 1-ethylbutyl, 2-ethylbutyl, 1,1,2-trimethylpropyl, 1,2,2-trimethylpropyl, 1-ethyl-1-methylpropyl and 1-ethyl-2-methylpropyl;

cycloalkyl: mono- or bicyclic saturated hydrocarbon groups having 3 to 6 carbon ring members, such as cyclopropyl, cyclobutyl, cyclopentyl and cyclohexyl.

alkoxyalkyl: a saturated straight-chain or mono-, di-, or tribranched hydrocarbon chain which is interrupted by an oxygen atom, for example C_2 - C_{11} -alkoxyalkyl: a hydrocarbon chain as described above having 2 to 11 carbon atoms which may be interrupted in any position by an oxygen atom, such as methoxyethyl, ethoxyethyl, propoxyethyl,

butoxyethyl, pentoxyethyl, hexyloxyethyl, heptyloxyethyl, octyloxyethyl, nonyloxyethyl, 3-(3-ethylhexyloxy)ethyl, 3-(2,4,4-trimethylpentyloxy)ethyl, 3-(1-ethyl-3-methylbutoxy)ethyl, methoxypropyl, ethoxypropyl, propoxypropyl, butoxypropyl,

pentoxypropyl, hexyloxypropyl, heptyloxypropyl, octyloxypropyl, nonyloxypropyl, 3-(3-ethylhexyloxy)propyl, 3-(2,4,4-trimethylpentyloxy)propyl, 3-(1-ethyl-3-methylbutoxy)propyl, methoxybutyl, ethoxybutyl, propoxybutyl, butoxybutyl, pentoxybutyl, hexyloxybutyl, octyloxybutyl, nonyloxybutyl, 3-(3-ethylhexyloxy)butyl, 3-(2,4,4-trimethylpentyloxy)butyl, 3-(1-ethyl-3-methylbutoxy)butyl, methoxypentyl, ethoxypentyl, propoxypentyl, butoxypentyl, pentoxypentyl, hexyloxypentyl.

The scope of the present invention includes the (R)- and (S)-isomers and the racemates of compounds of the formula I having chiral centers.

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With a view to the intended use of the triazolopyrimidines of the formula I, particular preference is given to the following meanings of the substituents, in each case on their own or in combination:

Preference is given to compounds I in which the sum of the carbon atoms in the groups R¹ and R² is at most 14.

The alkyl groups in \mathbb{R}^1 and \mathbb{R}^2 in formula I are preferably straight-chain or mono-, di- or tribranched alkyl groups.

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Preference is given to compounds I in which R¹ is methyl, ethyl, n-propyl, isopropyl, n-butyl or n-pentyl, in particular methyl or ethyl, where R¹ may be substituted as defined at the outset.

25 Preference is given to compounds I in which R¹ and R² which do not carry any substituents.

Particular preference is given to compounds I in which R^1 is C_5 - C_{12} -alkoxyalkyl, where the carbon chains are unsubstituted or may be substituted as defined at the outset.

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In one embodiment of the compounds I, R1 is alkoxyalkyl.

In another embodiment of the compounds I, both groups R^1 and R^2 are alkyl, which is substituted as defined at the outset or, preferably, unsubstituted.

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Preference is given to compounds I in which R^2 is a straight-chain or a mono- or dibranched C_5 - C_{12} -alkyl group which does not carry any further substituents.

In another embodiment of the compounds of the formula I, R^2 is branched at the α carbon atom. They are described by formula Ia:

$$\begin{array}{c}
N - N \\
N - N
\end{array}$$

$$\begin{array}{c}
R^1 \\
R^{21}
\end{array}$$

$$\begin{array}{c}
R^{21}
\end{array}$$

in which R^{21} is C_3 - C_{10} -alkyl or C_2 - C_{10} -alkenyl and R^{22} is C_1 - C_4 -alkyl, in particular methyl, where R¹¹ and R¹² together have at most 12 carbon atoms and are unsubstituted or may be substituted like R1 in formula I.

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If R1 or R2 contains a cyano group, this is preferably located at the terminal carbon atom.

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Particular preference is given to compounds I in which R² is n-pentyl, 1-methylbutyl, 2-methylbutyl, 3-methylbutyl, 2,2-dimethylpropyl, 1-ethylpropyl, n-hexyl, 1,1-dimethylpropyl, 1,2-dimethylpropyl, 1-methylpentyl, 2-methylpentyl, 3-methylpentyl, 4-methylpentyl, 1,1-dimethylbutyl, 1,2-dimethylbutyl, 1,3-dimethylbutyl, 2,2-dimethylbutyl, 2,3-dimethylbutyl, 3,3-dimethylbutyl, 1-ethylbutyl, 2-ethylbutyl, 1,1,2-trimethylpropyl, 1,2,2-trimethylpropyl, 1-ethyl-1-methylpropyl or 1-ethyl-2-methylpropyl.

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In a further preferred embodiment of the compounds of the formula I, R² is n-heptyl, 1-methylhexyl, n-octyl, 1-methylheptyl, n-nonyl, 1-methyloctyl, n-decyl, 1-methylnonyl, n-undecyl, 1-methyldecyl, n-dodecyl or 1-methylundecyl.

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In particular with a view to their use, preference is given to the compounds I compiled in the tables below. Moreover, the groups mentioned for a substituent in the tables are per se, independently of the combination in which they are mentioned, a particularly preferred embodiment of the substituent in question.

25 Table 1

Compounds of the formula I in which R1 is methyl and R2 for each compound corresponds to one row of Table A

Table 2

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Compounds of the formula I in which R1 is ethyl and R2 for each compound corresponds to one row of Table A

Table 3

Compounds of the formula I in which R1 is n-propyl and R2 for each compound corresponds to one row of Table A

Table 4

Compounds of the formula I in which R^1 is isopropyl and R^2 for each compound corresponds to one row of Table A

5 Table 5

Compounds of the formula I in which R¹ is n-butyl and R² for each compound corresponds to one row of Table A

Table 6

10 Compounds of the formula I in which R¹ is isobutyl and R² for each compound corresponds to one row of Table A

Table 7

Compounds of the formula I in which R¹ is sec-butyl and R² for each compound corresponds to one row of Table A

Table 8

Compounds of the formula I in which R^1 is n-pentyl and R^2 for each compound corresponds to one row of Table A

Table A

No.	No. R ²			
A-1	CH ₂ CH ₂ CH ₂ CH ₃			
A-2	CH(CH ₃)CH ₂ CH ₂ CH ₃			
A-3	CH ₂ CH(CH ₃)CH ₂ CH ₃			
A-4	CH ₂ CH ₂ CH(CH ₃)CH ₃			
A-5	CH ₂ CH ₂ CH(CH ₃) ₂			
A-6	CH(CH ₃)CH(CH ₃)CH ₃			
A-7	CH(CH ₃)CH(CH ₃) ₂			
A-8	· CH ₂ C(CH ₃) ₃			
A-9	CH ₂ CH ₂ CH ₂ CH ₂ CH ₃			
A-10	CH(CH ₃)CH ₂ CH ₂ CH ₃			
A-11	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃			
A-12	CH₂CH₂CH(CH₃)CH₂CH₃			
A-13	CH ₂ CH ₂ CH(CH ₃) ₂ CH ₂			
A-14	CH ₂ CH ₂ CH(CH ₃) ₂			
A-15	CH(CH ₃)CH(CH ₃)CH ₂ CH ₃			

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No.	R ²		
A-16	CH(CH ₃)CH ₂ CH(CH ₃) ₂		
A-17	CH₂CH₂C(CH₃)₃		
A-18	CH(CH ₃)CH ₂ CH(CH ₃)CH ₃		
A-19	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-20	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃		
A-21	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃		
A-22	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃		
A-23	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₃		
A-24	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₃		
A-25	CH₂CH₂CH₂CH(CH₃)₂		
A-26	CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₃		
A-27	CH₂CH(CH₃)CH(CH₃)CH₂CH₃		
A-28	CH ₂ CH ₂ CH ₂ C(CH ₃) ₃		
A-29	CH(CH ₃)CH ₂ CH(CH ₃)CH ₂ CH ₃		
A-30	CH₂CH(CH₃)CH(CH₃)CH₂CH₃		
A-31	CH(CH ₃)CH ₂ CH(CH ₃)CH ₃		
A-32	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-33	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-34	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃		
A-35	CH₂CH₂CH(CH₃)CH₂CH₂CH₃		
A-36	CH₂CH₂CH(CH₃)CH₂CH₃		
A-37	CH₂CH₂CH₂CH(CH₃)CH₂CH₃		
A-38	CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃) ₂		
A-39	CH ₂ CH ₂ CH ₂ C(CH ₃) ₃		
A-40	CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃		
A-41	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₃		
A-42	CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CH ₃		
A-43	CH(CH ₃)CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃		
A-44	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₃		
A-45	CH(CH₃)CH₂CH(CH₃)CH₂CH₃		
A-46	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃) ₂		
A-47	CH₂CH₂CH₂CH₂CH₂CH₂CH₃		

No.	R ²		
A-48	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-49	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-50	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-51	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃		
A-52	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃		
A-53	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ C(CH ₃) ₃		
A-54	CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-55	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃		
A-56	CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CH ₂ CH ₃		
A-57	CH(CH₃)CH₂CH(CH₃)CH₂CH₂CH₃		
A-58	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃		
A-59	CH(CH ₃)CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃		
A-60	CH(CH ₃)CH ₂ CH ₂ C(CH ₃) ₃		
A-61	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH(CH ₃) ₃		
A-62	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃) ₂		
A-63	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH(CH ₃) ₂		
A-64	CH ₂ CH ₃		
A-65	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-66	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-67	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-68	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂		
A-69	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃		
A-70	CH ₂ CH ₂ CH ₂ CH ₂ C(CH ₃) ₃ CH ₂		
A-71	CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-72	CH ₂ CH(CH ₃)CH(CH ₃) CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-73	CH ₂ CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-74	CH(CH ₃)CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-75	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-76	CH(CH ₃)CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃		
A-77	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃		
A-78	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₃		
A-79	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃) ₂		

No.	R ²		
A-80	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ C(CH ₃) CH ₃		
A-81	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃) CH ₃		
A-82	CH(CH ₃)CH ₂ CH ₂ CH ₂ C(CH ₃) ₃		
A-83	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ C(CH ₃) ₃		
A-84	CH ₂		
A-85	CH(CH ₃)CH ₂ CH ₃		
A-86	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-87	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-88	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-89	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-90	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ C(CH ₃) ₃		
A-91	CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-92	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-93	CH ₂ CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-94	CH(CH ₃)CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-95	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-96	CH(CH ₃)CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-97	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃		
A-98	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃		
A-99	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₃		
A-100	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃) ₂		
A-101	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₃		
A-102	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₃		
A-103	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃) ₂		
A-104	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ C(CH ₃) ₃		
A-105	CH ₂		
A-106	CH(CH ₃)CH ₂ CH ₂		
A-107	CH ₂ CH(CH ₃)CH ₂ CH ₃		
A-108	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-109	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂		
A-110	CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		
A-111	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃		

No.	R ²			
A-112	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃			
A-113	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃			
A-114	CH ₂			
A-115	CH ₂			
A-116	CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃			
A-117	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃			
A-118	CH ₂ CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃			
A-119	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃			
A-120	CH(CH ₃)CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃			
A-121	CH(CH ₃)CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₃			
A-122	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃			
A-123	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₃			
A-124	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃			
A-125	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₃			
A-126	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₃			
A-127	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₃			
A-128	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₃			
A-129	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ C(CH ₃) ₃			
A-130	CH ₂ CH ₂ CH ₂ CH ₂ CN			
A-131	CH(CH₃)CH₂CH₂CH2CN			
A-132	CH₂CH(CH₃)CH₂CH₂CN			
A-133	CH₂CH₂CH(CH₃)CH₂CN			
A-134	CH₂CH₂CH(CH₃)CH₂CN			
A-135	CH(CH₃)CH(CH₃)CH₂CN			
A-136	CH(CH₃)CH(CH₃)CH₂CN			

No.	R ²		
A-137	CH₂C(CH₃)₂CH₂CN		
A-138	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-139	CH(CH₃)CH₂CH₂CH₂CN		
A-140	CH₂CH(CH₃)CH₂CH₂CN		
A-141	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CN		
A-142	CH₂CH₂CH(CH₃)₂CH₂CH2CN		
A-143	CH₂CH₂CH₂CH(CH₃)CH₂CN		
A-144	CH(CH₃)CH(CH₃)CH₂CH		
A-145	CH(CH₃)CH₂CH(CH₃)CH₂CN		
A-146	CH₂CH₂C(CH₃)₂CH₂CN		
A-147	CH(CH₃)CH₂CH(CH₃)CH₂CN		
A-148	CH₂CH₂CH₂CH₂CH₂CH₂CN		
A-149	CH(CH₃)CH₂CH₂CH₂CH₂CN		
A-150	CH₂CH(CH₃)CH₂CH₂CH₂CH		
A-151	CH₂CH₂CH(CH₃)CH₂CH₂CH		
A-152	CH₂CH₂CH₂CH(CH₃)CH₂CH2CN		
A-153	CH₂CH₂CH₂CH(CH₃)CH₂CN		
A-154	CH₂CH₂CH₂CH(CH₃)CH₂CN		
A-155	CH(CH₃)CH(CH₃)CH₂CH₂CH		
A-156	CH₂CH(CH₃)CH(CH₃)CH₂CH₂CN		
A-157	CH₂CH₂CH2C(CH3)2CH2CN		
A-158	CH(CH₃)CH₂CH(CH₃)CH₂CH		
A-159	CH₂CH(CH₃)CH(CH₃)CH₂CH2CN		
A-160	CH(CH₃)CH₂CH₂CH(CH₃)CH₂CN		
A-161	CH₂CH₂CH₂CH₂CH₂CH₂CH₂CN		
A-162	CH(CH₃)CH₂CH₂CH₂CH₂CH₂CN		
A-163	CH₂CH(CH₃)CH₂CH₂CH₂CH₂CH		
A-164	CH₂CH₂CH(CH₃)CH₂CH₂CH₂CH₂CN		
A-165	CH₂CH₂CH2CH(CH₃)CH₂CH2CH2CN		
A-166	CH₂CH₂CH₂CH(CH₃)CH₂CH2CN		
A-167	CH₂CH₂CH₂CH₂CH(CH₃)CH₂CN		
A-168	CH ₂ CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CN		

No.	R ²		
A-169	CH(CH₃)CH(CH₃)CH₂CH₂CH₂CH2CN		
A-170	CH₂CH(CH₃)CH(CH₃)CH₂CH₂CH		
A-171	CH₂CH₂CH2C(CH3)2CH2CH2CN		
A-172	CH(CH₃)CH₂CH(CH₃)CH₂CH₂CH		
A-173	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CN		
A-174	CH(CH ₃)CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CN		
A-175	CH(CH ₃)CH ₂ CH ₂ CH(CH ₃)CH ₂ CN		
A-176	CH₂CH₂CH₂CH₂CH₂CH₂CH₂CH2CN		
A-177	CH(CH₃)CH₂CH₂CH₂CH₂CH₂CH₂CN		
A-178	CH₂CH(CH₃)CH₂CH₂CH₂CH₂CH₂CN		
A-179	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-180	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CN		
A-181	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CN		
A-182	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CN		
A-183	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-184	CH₂CH(CH₃)CH(CH₃)CH₂CH₂CH₂CH		
A-185	CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CH ₂ CN		
A-186	CH(CH₃)CH₂CH(CH₃)CH₂CH₂CH₂CN		
A-187	CH₂CH(CH₃)CH(CH₃)CH₂CH₂CH₂CH2CN		
A-188	CH(CH ₃)CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CN		
A-189	CH(CH₃)CH₂CH₂CH2C(CH₃)2CH2CN		
A-190	CH₂CH(CH₃)CH₂CH(CH₃)₂CH₂CN		
A-191	CH(CH₃)CH₂CH₂CH₂CH(CH₃)CH₂CN		
A-192	CH₂CH(CH₃)CH₂CH₂CH(CH₃)CH₂CN		
A-193	CH₂CH₂CH₂CH₂CH₂CH₂CH₂CH₂CH		
A-194	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-195	CH₂CH(CH₃)CH₂CH₂CH₂CH₂CH₂CH₂CH		
A-196	CH₂CH₂CH(CH₃)CH₂CH₂CH₂CH₂CH₂CH		
A-197	CH₂CH₂CH(CH₃)CH₂CH₂CH₂CH₂CH₂CN		
A-198	CH₂CH₂CH(CH₃)CH₂CH₂CH₂CH2CN		
A-199	CH ₂ CH ₂ CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CN		
A-200	CH(CH₃)CH(CH₃)CH₂CH₂CH₂CH₂CH₂CN		

No.	R ²		
A-201	CH ₂ CH(CH ₃)CH(CH ₃) CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-202	CH₂CH₂CH₂C(CH₃)₂CH₂CH₂CH₂CH2CN		
A-203	CH(CH₃)CH₂CH(CH₃)CH₂CH₂CH₂CH₂CH		
A-204	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-205	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-206	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-207	CH(CH₃)CH₂CH₂CH₂CH(CH₃)CH₂CH2CN		
A-208	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CN		
A-209	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ C(CH ₃)CH ₂ CN		
A-210	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CN		
A-211	CH(CH ₃)CH ₂ CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CN		
A-212	CH₂CH(CH₃)CH₂CH₂CH₂C(CH₃)₂CH₂CN		
A-213	CH2CH2CH2CH2CH2CH2CH2CH2CH2CH2CN		
A-214	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-215	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-216	CH2CH2CH(CH3)CH2CH2CH2CH2CH2CH2CH2CN		
A-217	CH2CH2CH2CH(CH3)CH2CH2CH2CH2CH2CH2CN		
A-218	CH2CH2CH2CH(CH3)CH2CH2CH2CH2CH2CN		
A-219	CH₂CH₂CH₂CH₂CH₂CH₂C(CH₃)₂CH₂CN		
A-220	CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-221	CH₂CH(CH₃)CH(CH₃)CH₂CH₂CH₂CH₂CH₂CH		
A-222	CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-223	CH(CH ₃)CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-224	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN		
A-225	CH(CH₃)CH₂CH(CH₃)CH₂CH₂CH₂CH₂CH		
A-226	CH(CH₃)CH₂CH₂CH₂CH(CH₃)CH₂CH₂CH₂CH		
A-227	CH(CH₃)CH₂CH₂CH₂CH(CH₃)CH₂CH₂CH		
A-228	CH(CH₃)CH₂CH₂CH₂CH₂CH(CH₃)CH₂CH2CN		
A-229	CH(CH₃)CH₂CH₂CH₂CH₂CH2CH(CH₃)CH2CN		
A-230	CH₂CH(CH₃)CH₂CH₂CH₂CH(CH₃)CH₂CH2CN		
A-231	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CN		
A-232	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CN		

No.	R²			
A-233	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CN			
A-234	CH2CH2CH2CH2CH2CH2CH2CH2CH2CH2CH2CH2CN			
A-235	CH(CH ₃)CH ₂ CH ₂			
A-236	CH2CH(CH3)CH2CH2CH2CH2CH2CH2CH2CH2CH2CN			
A-237	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN			
A-238	CH₂ CH₂CH(CH₃)CH₂CH₂CH₂CH₂CH₂CH₂CH2CN			
A-239	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN			
A-240	CH2CH2CH2CH2CH(CH3)CH2CH2CH2CH2CH2CN			
A-241	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH			
A-242	CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CN			
A-243	CH2CH2CH2CH2CH2CH2CH2CH(CH3)CH2CH2CN			
A-244	CH ₂			
A-245	CH(CH ₃)CH ₂ CH ₂			
A-246	CH ₂ CH(CH ₃)CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN			
A-247	CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN			
A-248	CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN			
A-249	CH(CH ₃)CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN			
A-250	CH(CH ₃)CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CN			
A-251	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CN			
A-252	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CN			
A-253	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CN			
A-254	CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₃)CH ₂ CH ₂ CN			
A-255	CH₂CH(CH₃)CH₂CH₂CH₂CH(CH₃)CH₂CH₂CH2CN			
A-256	CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CN			
A-257	CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ CN			
A-258	CH ₂ CH(CH ₃)CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ C(CH ₃) ₂ CH ₂ CN			

The compounds I are suitable as fungicides. They are distinguished by an outstanding effectiveness against a broad spectrum of phytopathogenic fungi, especially from the classes of the *Ascomycetes, Deuteromycetes, Oomycetes* and *Basidiomycetes*, especially from the class of the *Oomycetes*. Some are systemically effective and they can be used in plant protection as foliar fungicides, fungicides for seed-dressing and soil fungicides.

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They are particularly important in the control of a multitude of fungi on various cultivated plants, such as wheat, rye, barley, oats, rice, corn, grass, bananas, cotton, soya, coffee, sugar cane, grapevines, fruit and ornamental plants, and vegetables, such as cucumbers, beans, tomatoes, potatoes and cucurbits, and on the seeds of these plants.

They are especially suitable for controlling the following plant diseases:

- Alternaria species on fruit and vegetables,
- Bipolaris and Drechslera species on cereals, rice and lawns,
- Blumeria graminis (powdery mildew) on cereals,
 - Botrytis cinerea (gray mold) on strawberries, vegetables, ornamental plants and grapevines,
 - Bremia lactucae on lettuce
 - Erysiphe cichoracearum and Sphaerotheca fuliginea on cucurbits,
- Fusarium and Verticillium species on various plants,
 - Mycosphaerella species on cereals, bananas and peanuts,
 - Peronospora species on cabbage and bulbous plants,
 - Phakopsora pachyrhizi and P. meibomiae on soybeans,
 - Phytophthora infestans on potatoes and tomatoes,
- 20 Phytophthora capsici on peppers.
 - Plasmopara viticola on grapevines,
 - · Podosphaera leucotricha on apples,
 - Pseudocercosporella herpotrichoides on wheat and barley,
 - Pseudoperonospora species on hops and cucumbers,
- Puccinia species on cereals,
 - Pyricularia oryzae on rice,
 - Pythium aphanidermatum on lawns,
 - Rhizoctonia species on cotton, rice and lawns,
 - Septoria tritici and Stagonospora nodorum on wheat,
- 30 Uncinula necator on grapevines,
 - Ustilago species on cereals and sugar cane, and
 - Venturia species (scab) on apples and pears.

They are particularly suitable for controlling harmful fungi from the class of the

Oomycetes, such as Peronospora species, Phytophthora species, Plasmopara viticola and Pseudoperonospora species.

The compounds I are also suitable for controlling harmful fungi, such as *Paecilomyces variotii*, in the protection of materials (e.g. wood, paper, paint dispersions, fibers or fabrics) and in the protection of stored products.

The compounds I are employed by treating the fungi or the plants, seeds, materials or soil to be protected from fungal attack with a fungicidally effective amount of the active compounds. The application can be carried out both before and after the infection of the materials, plants or seeds by the fungi.

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The fungicidal compositions generally comprise between 0.1 and 95%, preferably between 0.5 and 90%, by weight of active compound.

When employed in plant protection, the amounts applied are, depending on the kind of effect desired, between 0.01 and 2.0 kg of active compound per ha.

In seed treatment, amounts of active compound of 1 to 1000 g/100 kg, preferably 5 to 100 g/100 kg of seed are generally required.

When used in the protection of materials or stored products, the amount of active compound applied depends on the kind of application area and on the desired effect. Amounts customarily applied in the protection of materials are, for example, 0.001 g to 2 kg, preferably 0.005 g to 1 kg, of active compound per cubic meter of treated material.

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The compounds I can be converted into the customary formulations, for example solutions, emulsions, suspensions, dusts, powders, pastes and granules. The application form depends on the particular intended use; in each case, it should ensure a fine and uniform distribution of the compound according to the invention.

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The formulations are prepared in a known manner, for example by extending the active compound with solvents and/or carriers, if desired using emulsifiers and dispersants. Solvents/auxiliaries which are suitable are essentially:

- water, aromatic solvents (for example Solvesso products, xylene), paraffins (for example mineral oil fractions), alcohols (for example methanol, butanol, pentanol, benzyl alcohol), ketones (for example cyclohexanone, gamma-butyrolactone), pyrrolidones (NMP, NOP), acetates (glycol diacetate), glycols, fatty acid dimethylamides, fatty acids and fatty acid esters. In principle, solvent mixtures may also be used.
- carriers such as ground natural minerals (for example kaolins, clays, talc, chalk) and ground synthetic minerals (for example highly disperse silica, silicates); emulsifiers such as nonionogenic and anionic emulsifiers (for example polyoxyethylene fatty alcohol ethers, alkylsulfonates and arylsulfonates) and dispersants such as lignosulfite waste liquors and methylcellulose.

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Suitable surfactants are alkali metal, alkaline earth metal and ammonium salts of lignosulfonic acid, naphthalenesulfonic acid, phenolsulfonic acid,

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dibutylnaphthalenesulfonic acid, alkylarylsulfonates, alkyl sulfates, alkylsulfonates, fatty alcohol sulfates, fatty acids and sulfated fatty alcohol glycol ethers, furthermore condensates of sulfonated naphthalene and naphthalene derivatives with formaldehyde, condensates of naphthalene or of naphthalenesulfonic acid with phenol and formaldehyde, polyoxyethylene octylphenol ether, ethoxylated isooctylphenol, octylphenol, nonylphenol, alkylphenol polyglycol ethers, tributylphenyl polyglycol ether, tristearylphenyl polyglycol ether, alkylaryl polyether alcohols, alcohol and fatty alcohol/ethylene oxide condensates, ethoxylated castor oil, polyoxyethylene alkyl ethers, ethoxylated polyoxypropylene, lauryl alcohol polyglycol ether acetal, sorbitol esters, lignosulfite waste liguors and methylcellulose.

Suitable for the preparation of directly sprayable solutions, emulsions, pastes or oil dispersions are mineral oil fractions of medium to high boiling point, such as kerosene or diesel oil, furthermore coal tar oils and oils of vegetable or animal origin, aliphatic, cyclic and aromatic hydrocarbons, for example toluene, xylene, paraffin, tetrahydronaphthalene, alkylated naphthalenes or their derivatives, methanol, ethanol, propanol, butanol, cyclohexanol, cyclohexanone, isophorone, strongly polar solvents, for example dimethyl sulfoxide, N-methylpyrrolidone and water.

Powders, materials for spreading and dustable products can be prepared by mixing or concomitantly grinding the active substances with a solid carrier.

Granules, for example coated granules, impregnated granules and homogeneous granules, can be prepared by binding the active compounds to solid carriers. Examples of solid carriers are mineral earths such as silica gels, silicates, talc, kaolin, attaclay, limestone, lime, chalk, bole, loess, clay, dolomite, diatomaceous earth, calcium sulfate, magnesium sulfate, magnesium oxide, ground synthetic materials, fertilizers, such as, for example, ammonium sulfate, ammonium phosphate, ammonium nitrate, ureas, and products of vegetable origin, such as cereal meal, tree bark meal, wood meal and nutshell meal, cellulose powders and other solid carriers.

In general, the formulations comprise from 0.01 to 95% by weight, preferably from 0.1 to 90% by weight, of the active compound. The active compounds are employed in a purity of from 90% to 100%, preferably 95% to 100% (according to NMR spectrum).

The following are examples of formulations: 1. Products for dilution with water

A Water-soluble concentrates (SL)

10 parts by weight of a compound according to the invention are dissolved in water or in a water-soluble solvent. As an alternative, wetters or other auxiliaries are added. The active compound dissolves upon dilution with water.

B Dispersible concentrates (DC)

20 parts by weight of a compound according to the invention are dissolved in cyclohexanone with addition of a dispersant, for example polyvinylpyrrolidone. Dilution with water gives a dispersion.

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C Emulsifiable concentrates (EC)

15 parts by weight of a compound according to the invention are dissolved in xylene with addition of calcium dodecylbenzenesulfonate and castor oil ethoxylate (in each case 5%). Dilution with water gives an emulsion.

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D Emulsions (EW, EO)

40 parts by weight of a compound according to the invention are dissolved in xylene with addition of calcium dodecylbenzenesulfonate and castor oil ethoxylate (in each case 5%). This mixture is introduced into water by means of an emulsifying machine (Ultraturrax) and made into a homogeneous emulsion. Dilution with water gives an emulsion.

E Suspensions (SC, OD)

In an agitated ball mill, 20 parts by weight of a compound according to the invention are comminuted with addition of dispersants, wetters and water or an organic solvent to give a fine active compound suspension. Dilution with water gives a stable suspension of the active compound.

F Water-dispersible granules and water-soluble granules (WG, SG)

50 parts by weight of a compound according to the invention are ground finely with addition of dispersants and wetters and made into water-dispersible or water-soluble granules by means of technical appliances (for example extrusion, spray tower, fluidized bed). Dilution with water gives a stable dispersion or solution of the active compound.

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G Water-dispersible powders and water-soluble powders (WP, SP)
75 parts by weight of a compound according to the invention are ground in a rotor—
stator mill with addition of dispersants, wetters and silica gel. Dilution with water gives a
stable dispersion or solution of the active compound.

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2. Products to be applied undiluted

H Dustable powders (DP)

5 parts by weight of a compound according to the invention are ground finely and 40 mixed intimately with 95% of finely divided kaolin. This gives a dustable product. I Granules (GR, FG, GG, MG)

0.5 part by weight of a compound according to the invention is ground finely and associated with 95.5% carriers. Current methods are extrusion, spray-drying or the fluidized bed. This gives granules to be applied undiluted.

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J ULV solutions (UL)

10 parts by weight of a compound according to the invention are dissolved in an organic solvent, for example xylene. This gives a product to be applied undiluted.

The active compounds can be used as such, in the form of their formulations or the use forms prepared therefrom, for example in the form of directly sprayable solutions, powders, suspensions or dispersions, emulsions, oil dispersions, pastes, dustable products, materials for spreading, or granules, by means of spraying, atomizing, dusting, spreading or pouring. The use forms depend entirely on the intended uses; the intention is to ensure in each case the finest possible distribution of the active compounds according to the invention.

Aqueous use forms can be prepared from emulsion concentrates, pastes or wettable powders (sprayable powders, oil dispersions) by adding water. To prepare emulsions, pastes or oil dispersions, the substances, as such or dissolved in an oil or solvent, can be homogenized in water by means of a wetter, tackifier, dispersant or emulsifier. Alternatively, it is possible to prepare concentrates composed of active substance, wetter, tackifier, dispersant or emulsifier and, if appropriate, solvent or oil, and such concentrates are suitable for dilution with water.

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The active compound concentrations in the ready-to-use preparations can be varied within relatively wide ranges. In general, they are from 0.0001 to 10%, preferably from 0.01 to 1%.

The active compounds may also be used successfully in the ultra-low-volume process (ULV), by which it is possible to apply formulations comprising over 95% by weight of active compound, or even to apply the active compound without additives.

Various types of oils, wetters, adjuvants, herbicides, fungicides, other pesticides, or bactericides may be added to the active compounds, if appropriate not until immediately prior to use (tank mix). These agents can be admixed with the agents according to the invention in a weight ratio of 1:10 to 10:1.

The compositions according to the invention can, in the use form as fungicides, also be present together with other active compounds, e.g. with herbicides, insecticides, growth regulators, fungicides or else with fertilizers. Mixing the compounds I or the compositions comprising them in the application form as fungicides with other

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fungicides results in many cases in an expansion of the fungicidal spectrum of activity being obtained.

The following list of fungicides, in conjunction with which the compounds according to the invention can be used, is intended to illustrate the possible combinations but does not limit them:

- acylalanines, such as benalaxyl, metalaxyl, ofurace or oxadixyl,
- amine derivatives, such as aldimorph, dodine, dodemorph, fenpropimorph, fenpropidin, guazatine, iminoctadine, spiroxamine or tridemorph,
 - anilinopyrimidines, such as pyrimethanil, mepanipyrim or cyprodinyl,
- antibiotics, such as cycloheximide, griseofulvin, kasugamycin, natamycin, polyoxin or streptomycin,
- azoles, such as bitertanol, bromoconazole, cyproconazole, difenoconazole, dinitroconazole, enilconazole, epoxiconazole, fenbuconazole, fluquinconazole, flusilazole, hexaconazole, imazalil, metconazole, myclobutanil, penconazole, propiconazole, prochloraz, prothioconazole, tebuconazole, triadimefon, triadimenol, triflumizole or triticonazole,
 - dicarboximides, such as iprodione, myclozolin, procymidone or vinclozolin,
- dithiocarbamates, such as ferbam, nabam, maneb, mancozeb, metam, metiram, propineb, polycarbamate, thiram, ziram or zineb,
 - heterocyclic compounds, such as anilazine, benomyl, boscalid, carbendazim, carboxin, oxycarboxin, cyazofamid, dazomet, dithianon, famoxadone, fenamidone, fenarimol, fuberidazole, flutolanil, furametpyr, isoprothiolane, mepronil, nuarimol, probenazole, proquinazid, pyrifenox, pyroquilon, quinoxyfen, silthiofam, thiabendazole, thifluzamide, thiophanate-methyl, tiadinil, tricyclazole or triforine,
 - copper fungicides, such as Bordeaux mixture, copper acetate, copper oxychloride or basic copper sulfate,
 - nitrophenyl derivatives, such as binapacryl, dinocap, dinobuton or nitrophthalisopropyl,
 - · phenylpyrroles, such as fenpicionil or fludioxonil,
 - sulfur,
- other fungicides, such as acibenzolar-S-methyl, benthiavalicarb, carpropamid, chlorothalonil, cyflufenamid, cymoxanil, diclomezine, diclocymet, diethofencarb, edifenphos, ethaboxam, fenhexamid, fentin acetate, fenoxanil, ferimzone, fluazinam, phosphorous acid, fosetyl, fosetyl-aluminum, iprovalicarb, hexachlorobenzene, metrafenone, pencycuron, propamocarb, phthalide, tolclofosmethyl, quintozene or zoxamide,
- strobilurins, such as azoxystrobin, dimoxystrobin, enestroburin, fluoxastrobin,
 kresoxim-methyl, metominostrobin, orysastrobin, picoxystrobin, pyraclostrobin or trifloxystrobin,

- sulfenic acid derivatives, such as captafol, captan, dichlofluanid, folpet or tolylfluanid,
- cinnamides and analogous compounds, such as dimethomorph, flumetover or flumorph.

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Synthesis examples

The procedures illustrated in the synthesis examples below were, with appropriate modification of the starting materials, used to obtain further compounds I. The compounds obtained in this manner are listed in the table that follows, together with physical data.

Example 1: Synthesis of 1-methyl-2-oxooctane-1-nitrile

At -75°C, 100 ml of a 15% by weight strength solution of butyl lithium in hexane were added, over a period of 1 hour, to a solution of 8.25 g (0.15 mol) of propionitrile in 200 ml of anhydrous tetrahydrofuran (THF). The temperature was maintained, and after 2 hours of stirring, 21.6 g (0.15 mol) of methyl heptanoate were added dropwise over a period of 1 hour. After a further 2 hours of stirring, the solution was allowed to warm to about 20-25°C over about 14 hours. At 5-10°C, 50 ml of water were added to the reaction mixture and the pH was adjusted to 1-2 using semi-concentrated hydrochloric acid. The organic phase was removed and washed with water and dilute aqueous NaHCO₃ solution until neutral. After drying, the solvent was distilled off. This gave 26.0 g of the title compound as a brown oil.

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Example 2: Preparation of 5-hexyl-6-methyl-7-aminotriazolopyrimidine [1-2]

At 190°C, a suspension of 10.0 g (60 mmol) of the nitrile from Ex. 1, 3.8 g (45 mmol) of 3-amino-1,2,4-triazole and 1.7 g (9 mmol) of p-toluenesulfonic acid in 50 ml of mesitylene was heated on a water separator for 4 hours. The mesitylene was then distilled off, and the residue was digested with dichloromethane/water. The residue was filtered off, dried and chromatographed on silica gel using dichloromethane/ethyl acetate. This gives 0.5 g of the title compound in the form of colorless crystals.

Table I - Compounds of the formula I

No.	R ¹	R ²	Phys. Data (m.p. [°C])
I-1	CH ₃	(CH ₂) ₄ CH ₃	222-223
I-2	CH₃	(CH ₂)₅CH ₃	193-194
I-3	CH₃	(CH ₂) ₆ CH ₃	182-183
I-4	CH₃	(CH ₂) ₇ CH ₃	191-192
I-5	CH₃	(CH ₂) ₈ CH ₃	181-182
I-6	CH₃	(CH ₂) ₉ CH ₃	187-188
1-7	CH₂CH₃	(CH ₂) ₇ CH ₃	184-185
1-8	CH₂CH₃	(CH ₂) ₈ CH ₃	178-179
I-9	CH₂CH₃	(CH ₂) ₉ CH ₃	180-181
I-10	CH₂CH₂CH₃	(CH ₂) ₇ CH ₃	162-163
I-11	CH₂CH₂CH₃	(CH ₂) ₈ CH ₃	161-162
I-12	CH₂CH₂CH₃	(CH ₂) ₉ CH ₃	151-152
I-13	CH ₂ CH ₂ CH ₂ CH ₃	(CH₂) ₆ CH ₃	152-156

5 Examples of the action against harmful fungi

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The fungicidal action of the compounds of the formula I was demonstrated by the following experiments:

The active compounds were prepared as a stock solution comprising 25 mg of active compound which was made up to 10 ml using a mixture of acetone and/or DMSO and the emulsifier Uniperol® EL 9 (wetting agent having emulsifying and dispersing action based on ethoxylated alkyl phenols) in a ratio by volume of solvent:emulsifier of 99:1. The mixture was then made up to 100 ml with water. This stock solution was diluted with the solvent/emulsifier/water mixture described to the active compound concentration stated below.

Use Example 1: Activity against late blight of tomatoes caused by *Phytophthora infestans*, protective treatment

Leaves of potted tomato plants were sprayed to runoff point with an aqueous suspension of the active compounds. The next day, the leaves were infected with an aqueous sporangia suspension of *Phytophthora infestans*. The plants were then placed on a

water-vapor-saturated chamber at temperatures between 18 and 20°C. After 6 days the late blight on the untreated, but infected control plants had developed to such an extent that the infection could be determined visually in %.

In this test, the plants which had been treated with 250 ppm of the compound I-1 or I-2 showed an infection of at most 10%, whereas the untreated plants were 90% infected.

Use Example 2 – Activity against peronospora of grapevines caused by *Plasmopara viticola* 5 day protective application

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Leaves of potted vines were sprayed to runoff point with an aqueous suspension having the concentration of active compounds stated below. Five days after application, the undersides of the leaves were inoculated with an aqueous sporangia suspension of *Plasmopara viticola*. The vines were then initially placed in a water-vapor-saturated chamber at 24°C for 48 hours and then in a greenhouse at temperatures between 20°C and 30°C for five days. After this time, the plants were once more placed in a humid chamber for 16 hours to promote the eruption of sporangiophores. The extent of the development of the infection on the undersides of the leaves was then determined visually.

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In this test, the plants which had been treated with 250 ppm of the active compound I-1, I-2, I-10 or I-13 showed an infection of at most 10%, whereas the untreated plants were 80% infected.